

INTERACTION OF RADIUM WITH FRESHWATER SEDIMENTS AND THEIR MINERAL COMPONENTS

II. KAOLINITE AND MONTMORILLONITE

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Radiotracer method has been used for investigation of the adsorption and desorption of traces of radium on kaolinite and montmorillonite under conditions similar to those prevailing in waste and surface waters. It has been found that adsorption depends on the pH and ionic strength of the solution, and the character of these dependences is different for kaolinite and montmorillonite. Sulfates at concentration $60 \text{ mg} \cdot \text{l}^{-1}$ have negligible effect on the adsorption. Montmorillonite represents better adsorbent for radium than kaolinite, but both minerals may significantly affect the form and migration of radium in surface waters, if present as (major) components of freshwater sediments. Radium can be quantitatively desorbed by 1M HCl from kaolinite but not from montmorillonite. The desorption by 0.1M NaCl is generally lower and depends on the pH during the preliminary adsorption. Mechanisms of radium adsorption on both minerals and the character of the adsorption sites are discussed.

Introduction

Knowledge of the mechanisms and laws of interaction of radium with freshwater sediments is rather scarce, although the interaction substantially influences environmental migration of radium released from the nuclear fuel cycle.¹ Therefore, we began² a study of radium interaction with model solids representing mineral components of the sediments. Our approach consists in the analysis of the effects of the most important variables on the distribution of radium between the solids and water phase under conditions similar to those encountered in waste and natural waters. The final goal is to assess the significance of the individual model solids for radium migration in surface waters. This can be achieved also by comparison of the data obtained for the model solids with those characterizing radium interaction with sediments containing these solids.

The first two model solids chosen for the study were ferric hydroxide and quartz.² This paper presents the data on radium interaction with kaolinite and montmorillonite, which are minerals most often encountered in stream sediments and common in soils. Because of their high adsorption capacity and large active surface in dispersed state, the minerals hinder the migration of radionuclides and in the form of compact sediments may serve as an effective barrier against radionuclide migration also due to their low permeability.³

Experimental

Adsorbents

Bentonite from Ginovci, Yugoslavia, denoted as "rubber bentonite", served as the sample of montmorillonite. It contained 95% monoclinic montmorillonite, 3–5% illite and accessories of quartz. It had good mechanical properties, chemical and thermal stability and high cation exchange capacity (CEC = 90.8 meq/100 g). Total specific surface area of the sample was 750 m²/g as determined using the ethylene glycol monoethyl ether method, of this value 672 m²/g belonged to inter-layer spaces. Crystallochemical density of the sample was 2.735 g/cm³, value of the tetrahedral twist was 7°54', charge density 11.71 μC/m² and parameters of crystal lattice a = 0.518 nm, b = 0.898 nm. Due to the isomorphous substitution of atoms in the structure of montmorillonite the largest charge deficit was in octahedral sheets (−0.837), and smaller in tetrahedral ones (−0.088). The structural crystallochemical formula of montmorillonite was (Si_{7.912} Al_{0.088}) (Al_{3.018} Fe³⁺_{0.149} Fe²⁺_{0.033} Mn_{0.004} Mg_{0.800}) (Mg_{0.274} Ca_{0.172} Na_{0.008} K_{0.013}) O₂₀ (OH)₄.

Kaolinite was represented by washed kaolin S1a from locality Božičany near Karlovy Vary, Czechoslovakia. The kaolin contained 93–95% kaolinite with well ordered structure, with crystallinity index I = 0.84 (after HINCKLEY⁴), and 4% muscovite and 2% quartz. Specific surface area of the sample, determined by the low temperature (liquid N₂) adsorption of nitrogen from the mixture H₂ : N₂ = 3:1 was 13.42 m²/g, CEC measured was 6.2 meq/100 g and CEC calculated was 3.8 meq/100 g. The crystal lattice parameters of triclinic kaolinite are: a = 0.515 nm, b = 0.894 nm, c = 0.739 nm, α = 91.8°, β = 104.6°, γ = 90°; the crystallochemical density is 2.616 g/cm³. The surface charge density was 44.58 μC/cm², value of the tetrahedral twist was 8°59'. The probable crystallochemical formula of the kaolinite was



as compared with the ideal formula $\text{Si}_4\text{Al}_4\text{O}_{10}(\text{OH})_8$. The difference can be ascribed to the presence in kaolinite of impurities other than muscovite and quartz.

The studies were carried out with fractions of bentonite and kaolin containing grains in the range 0.2–0.05 μm , obtained by sieving. However, the prevailing particle sizes after dispersion in aqueous medium were $<1 \mu\text{m}$ e.s.d. for montmorillonite and $<2 \mu\text{m}$ e.s.d. for kaolinite. Desired amount of adsorbent was weighed dry into a 30 ml polythene bottle where the experiment was carried out.

Aqueous phase

Experimental solutions were prepared from distilled water and p.a. reagents. They were labelled with ^{224}Ra (carrier-free, $T = 3.64 \text{ d}$), obtained by milking of a generator⁵ with 0.01M HCl. Radium concentration in the experimental solution was 8–40 pg/l, the variation exerted no influence on the results. For other details see Ref. 2.

Procedures and evaluation

Adsorbent was shaken with 10 ml of experimental solution at room temperature for 17 hrs. After separation of the adsorbent and aqueous phase by centrifugation (3000 r.p.m., 10 min), the concentration of radium remaining in the solution (represented by radioactivity X) was determined by measurement of gamma radiation of the solution using well-type NaI(Tl) crystal and nuclear counter NZQ 714T (Tesla, Přemyslení, Czechoslovakia) and pH of the solution was determined with a glass electrode G 202 C and pH-meter PHM-52 (Radiometer, Copenhagen). The empty polythene bottle was shaken for 3 hrs with 10 ml 1M HCl, the radioactivity Y of the resulting solution represented the amount of radium adsorbed on the walls of the bottle during adsorption experiment. The percentage of radium so adsorbed was calculated as $100 Y S^{-1}$, where S is radioactivity of the same volume of experimental solution prior to adsorption. It seldom exceeded 5% of the total amount of radium present. The percentage of radium (A) adsorbed on clay adsorbents was calculated from

$$A = 100 (S - X - Y) (S - Y)^{-1}.$$

In order to study the desorption of radium from clay, 10 ml of desorption solution was added to clay after the adsorption, centrifugation and removal of adsorption solu-

tion. After shaking at room temperature for 17 hrs, the solid phase was again separated by centrifugation and radioactivity Z of the desorption solution was measured. Desorption percentage D was then calculated from $D = 100 Z (S-X)^{-1}$, or reversibility of the adsorption was expressed as R (in %) = $100 D (100-A)$. Errors due to incomplete removal of adsorption solution from adsorbent prior to desorption were lower than 2%. Further details of the procedures can be found in Ref. 2.

Some of the results obtained were expressed using distribution coefficients K_D and K'_D defined as

$$K_D = \frac{A}{100-A} \cdot \frac{V}{m},$$

and (see Ref. 6.)
$$K'_D = \frac{A}{A_{\max}-A} \cdot \frac{V}{m}.$$

Here m is the amount of adsorbent in g, V is the volume of adsorption solution (10 cm^3), A is the adsorption percentage found at the given V/m and $A_{\max}$ is maximum adsorption percentage for the given adsorbent, which cannot be exceeded at any increase in m/V .

Results and discussion

Adsorption of radium was studied as a function of several variables, whose changes might affect the behaviour of radium in waste and natural waters. Conditions of the experiments were chosen so as to simulate as closely as possible the conditions prevailing in real systems.²

The results of experiments are presented in this paper as points in graphs or numerical values in tables. Each point represents single experiment, the numerical values are averages from 2–4 experiments.

Preliminary experiments have shown that the kinetics of radium adsorption on kaolinite and montmorillonite is very rapid. The adsorbed amount does not change from 1 hr to 17 hr contact, most of radium is adsorbed during the first 10 min of shaking. It can therefore be assumed that the adsorption values resulting from 17 hr experiments represent equilibrium (or pseudoequilibrium, if the adsorption is not completely reversible) values of radium distribution. Furthermore, kinetics of radium adsorption on kaolinite and montmorillonite in natural water systems will be more probably determined by transport of radium to the surface of particles (diffusion, eddy diffusion) than by the adsorption process itself.

Kaolinite

Figs 1 and 2 show the dependence of radium adsorption by kaolinite on the pH and composition of adsorption solution and on the liquid to solid ratio, V/m . The S-shaped course of the pH dependences is similar to the course of radium adsorption on quartz,² but the pH dependence is less pronounced than that on quartz and radium is significantly adsorbed even at low pH values. The pH dependence of radium adsorption by kaolinite can be explained by the competition of H^+ ions with Ra^{2+} for the same adsorption sites and by the effect of pH on the sorption properties of kaolinite. This effect is due to several reasons.

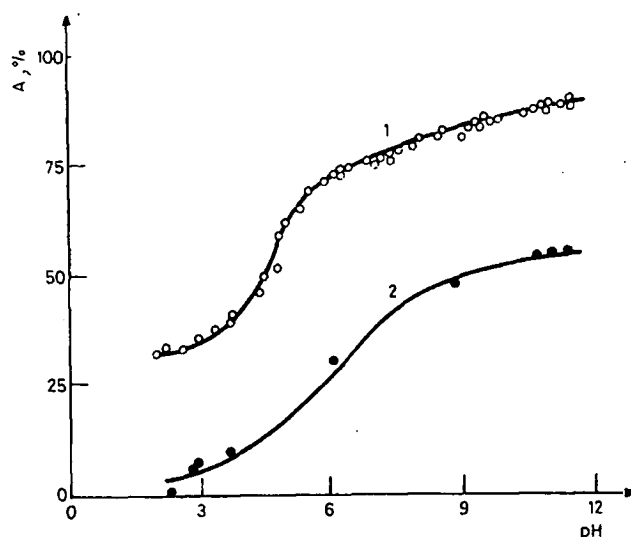


Fig. 1. Adsorption of radium (in %) on kaolinite from aqueous solutions of $0.01M Na^+$ as a function of pH of the solutions and liquid to solid ratio (V/m). Curve 1 – $V/m = 500 \text{ cm}^3 \cdot \text{g}^{-1}$; curve 2 – $V/m = 5000 \text{ cm}^3 \cdot \text{g}^{-1}$

Edges of the tetrahedral and octahedral sheets of kaolinite distorted by mechanical crushing are carriers of negatively or positively charged sites. The crushing leads to the rupture of $-M-O-M-$ bonds in the sheets (M denotes Al or Si). Oxygen atom in the ruptured bond retains the bond electrons and, consequently, two oppositely charged groups are formed by the rupture: $-M-\bar{O}$ and $^+M-O-$. The negatively charged group easily binds proton from acidic solution and becomes neutral. The neutralization depends on pH. With decreasing pH, the groups located on the edges of the octahedral sheets ($-Al-O^-$, $-Fe-O^-$, $-Mg-O^-$) are first neutralized, followed by the groups on the edges of the tetrahedral sheets ($-Si-O^-$) at still lower pH. Consequently, the amount of negatively charged

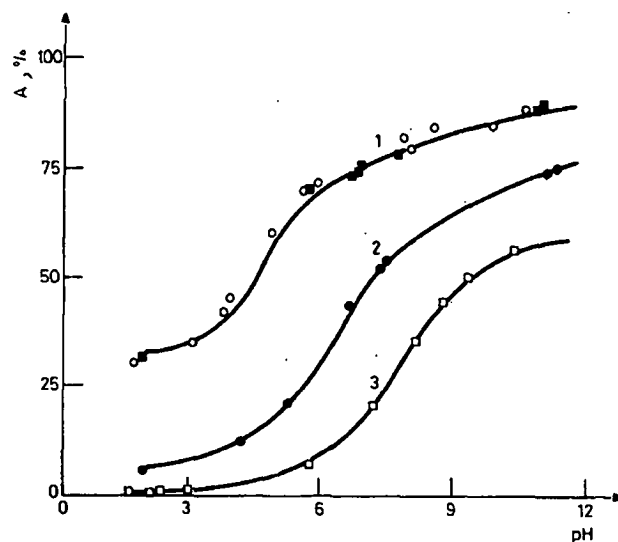


Fig. 2. Adsorption of radium (in %) on kaolinite ($V/m = 500 \text{ cm}^3 \cdot \text{g}^{-1}$) as a function of pH and composition of solution. Curves: 1 – 0.001M Na^+ ($\circ$) and 0.01M Na^+ , $60 \text{ mg} \cdot \text{l}^{-1} \text{SO}_4^{2-}$ ($\blacksquare$); 2 – 0.01M Na^+ , $30 \text{ mg} \cdot \text{l}^{-1} \text{Ca}^{2+}$; 3 – 0.1M NaCl

sites capable of binding radium diminishes, which is reflected in the decrease of radium adsorption. This effect can also be interpreted as competition between H^+ and Ra^{2+} ions from which it is indistinguishable.

The positively charged $-\text{O}-\text{M}^+$ groups are inactive towards cations in acidic solutions. They can, however, bind hydroxide ions from solution and the surface groups $-\text{Si}-\text{OH}$ (in tetrahedral sheets) or $-\text{Al}-\text{OH}$ (in octahedral sheets) thus formed can dissociate in slightly acidic, neutral or alkaline medium, releasing H^+ cation⁷ and forming negatively charged adsorption sites for radium. This process is supported by increasing pH and leads to an increase in the adsorption of radium with increasing pH.

The effects just discussed can be relevant particularly in the pH region of 4–11. In strongly acidic solutions, still other effects can be considered. At pH less than 3, kaolinite significantly decomposes under the release of Al^{3+} ions from octahedra of the crystal lattice.⁸ Al^{3+} ions thus released are partially bound at the surface exchange sites of the crystals. Strength of the bond of the ions is much larger than that of radium and therefore the dissolution may bring about a decrease in radium adsorption. Another phenomenon relevant for radium adsorption is the agglomeration of kaolinite particles which is known to occur at low (<4) pH values. Individual kaolinite crystals are united in their oppositely charged sites (negatively charged faces with positively charged edges of the crystals) and a “card-house” structure is formed. This process is facilitated by the suitable ratio

of the length to the thickness of the plate-like particles of kaolinite, which is about 10:1. The agglomeration leads to the reduction of surface area available for ion adsorption. However, neither the dissolution nor the agglomeration seem to be manifested in the pH dependences of radium adsorption shown in Figs 1 and 2.

The most interesting feature of the pH dependence is the substantial adsorption of radium on kaolinite at pH less than 4, where analogous adsorption of radium on quartz is very low.² The difference cannot be due to the presence of $-Al-O^-$ sites on kaolinite, because these sites are active at higher pH values than $-Si-O^-$ sites. We believe that radium is adsorbed at pH < 4 predominantly electrostatically due to the action of negative charges formed in kaolinite by the substitution of Mg^{2+} for Al^{3+} in octahedral sheets. The total charge of kaolinite formed by the substitution is rather small but sufficient for the adsorption of radium present at such low concentrations. The charge is independent of pH and, consequently, the pH will exert a small (if any) effect on this kind of radium adsorption. However, the surface charge of kaolinite formed in this way may strongly depend on the extent and quality of the substitution so that radium adsorption will probably be affected by the degree of structural ordering of layers in the crystal structure of kaolinite, by the amount of admixtures, etc. Another important effect possible in nature may be due to the surface coating of kaolinite particles with hydrated oxides of Fe(III).⁹ Such a coating would decrease the surface area of kaolinite available for radium adsorption, or it would change the mechanism of the adsorption.

As can be seen from Fig. 2, the addition to the adsorption solution of sulfates at a concentration typical for river waters does not change radium adsorption on kaolinite. Curve 1 in Fig. 2 is identical with curve 1 in Fig. 1. Similarly, no difference could be found when the concentration of sodium ions in the solution was changed from 0.01M to 0.001M. Sodium ions at such low concentrations prevent agglomeration of the clay and do not compete significantly with radium in the adsorption. The suppression of radium adsorption by the increase of sodium ion concentration to 0.1M and by the addition of Ca^{2+} ions at a concentration typical for river waters (Fig. 2) can be ascribed to the competition effect of these ions and/or to the agglomeration of kaolinite particles, the former effect being more probable.

Comparison of curves 1 and 2 in Fig. 1 shows that the adsorption of radium on kaolinite decreases with increasing liquid to solid ratio (V/m) in all the pH region studied. This effect was investigated in more detail at pH 7.58 and the results are presented in Table 1. It has been found that the adsorption increases with decreasing V/m to the maximum value of 95.2% at $V/m \geq 20 \text{ cm}^3 \cdot \text{g}^{-1}$. The value of K_D decreases with decreasing V/m and therefore K_D is not suitable for quantitative description of radium adsorption on kaolinite, although it is often used for various purposes (see for instance Ref.¹). The modified distribution coefficient, K'_D , is

Table 1
Adsorption (A) of radium on kaolinite from the
solution of pH 7.58 (0.005M Na₂B₄O₇, 0.005M HCl) as
a function of liquid to solid ratio (V/m)

V/m, cm ³ · g ⁻¹	A, %	K _D , cm ³ · g ⁻¹	K' _D , cm ³ · g ⁻¹
5000	39.1	3210	3485
2500	52.0	2708	3009
500	79.2	1904	2475
400	82.5	1885	2598
200	89.5	1705	3140
100	91.6	1090	2544
50	93.7	744	3123
40	93.8	605	2680
25	94.5	430	3375
20	95.2	397	—
10	95.2	198	—

$$\text{Mean } K'_D = 2937 \pm 125 \text{ cm}^3 \cdot \text{g}^{-1}$$

fairly constant and can be used for quantitative characterization of the adsorption. Similar conclusions were drawn earlier for other systems,² but a suitable interpretation of the non-constancy of K_D and of the limit for the adsorption lower than 100% was not yet proposed.

It can be concluded from the above data that kaolinite may represent an important component of suspended or bottom freshwater sediments from the point of view of radium adsorption. Its importance will depend on the pH value and composition of aqueous phase and on its abundance in the sediments, but signi-

Table 2
Desorption (D) of radium from kaolinite by 0.1M NaCl and 1M HCl
as a function of pH during the preliminary adsorption, and
reversibility (R) of the adsorption

pH	D, % 0.1M NaCl	pH	D, % 1M HCl	pH	R, %
2.1	88.5	2.5	100	2.1	52.7
5.6	80.2	4.5	100	3.2	66.1
6.1	79.3	6.6	100	5.4	68.8
7.3	73.9	7.4	100	6.4	69.2
7.8	74.6	9.2	99.8	6.8	64.1
8.5	73.5	11.3	99.9	7.8	63.2
9.2	69.1			9.2	65.2
11.2	70.2			11.2	64.5

ficant adsorption of radium can occur on the kaolinite component even at rather low pH values (< 6). The importance of kaolinite may decrease due to surface coating with other materials.

Table 2 contains data on radium desorption from kaolinite. The preliminary adsorption took place on 20 mg kaolinite from a solution with ionic strength equal to 0.01M. From Table 2 it follows that radium can be quantitatively desorbed with 1M HCl.

The desorption is rather high also with 0.1M NaCl, but moderately decreases with increasing pH at preliminary adsorption. The adsorption is quite reversible, the reversibility is virtually independent of the pH value of the solution. These findings indicate rather weak bonding of radium on the surface of kaolinite, whose strength does not change substantially with pH. Consequently, the adsorption bonds formed at different pH values with different surface groups have about the same strength. Radium adsorbed on the kaolinite components of freshwater sediments can be easily released under changed ambient conditions.

Montmorillonite

The course of radium adsorption on montmorillonite is shown in Figs 3 and 4 as a function of pH and composition of the adsorption solution. From the comparison of Fig. 3 with Fig. 1, it follows that the pH-dependence of radium

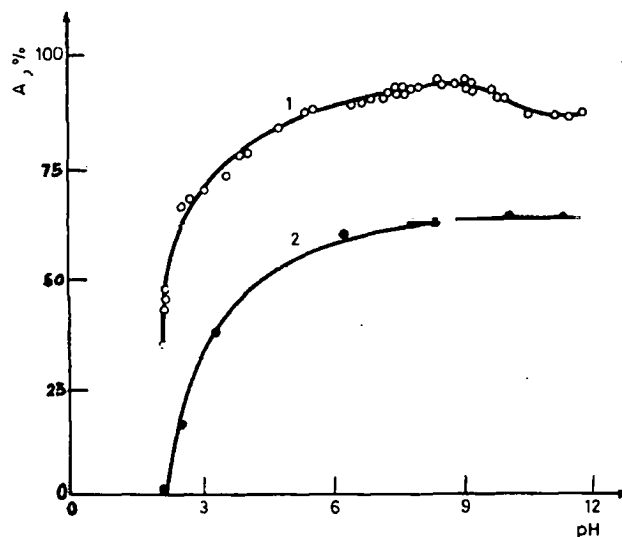


Fig. 3. Adsorption of radium (in %) on montmorillonite from aqueous solutions of 0.01M Na⁺ as a function of pH of the solution and liquid to solid ratio (V/m). Curve 1 – V/m = 500 cm³ · g⁻¹; curve 2 – V/m = 5000 cm³ · g⁻¹

Table 3
Adsorption of radium (A) on montmorillonite from the
solution of pH 7.47 (0.005M $\text{Na}_2\text{B}_4\text{O}_7$, 0.005M HCl) as
a function of liquid to solid ratio (V/m)

V/m, $\text{cm}^3 \cdot \text{g}^{-1}$	A, %	K_D , $\text{cm}^3 \cdot \text{g}^{-1}$	K'_D , $\text{cm}^3 \cdot \text{g}^{-1}$
5000	61.9	8123	8456
3333	70.2	7852	8268
2000	79.5	7756	8368
1000	88.9	8009	9260
500	92.9	6542	8295
400	94.1	6380	8555
200	96.0	4800	7680
100	97.5	3900	9750
50	98.0	2450	9800
25	98.5	1641	—
10	98.5	657	—

$$\text{Mean } K'_D = 8716 \pm 241 \text{ cm}^3 \cdot \text{g}^{-1}$$

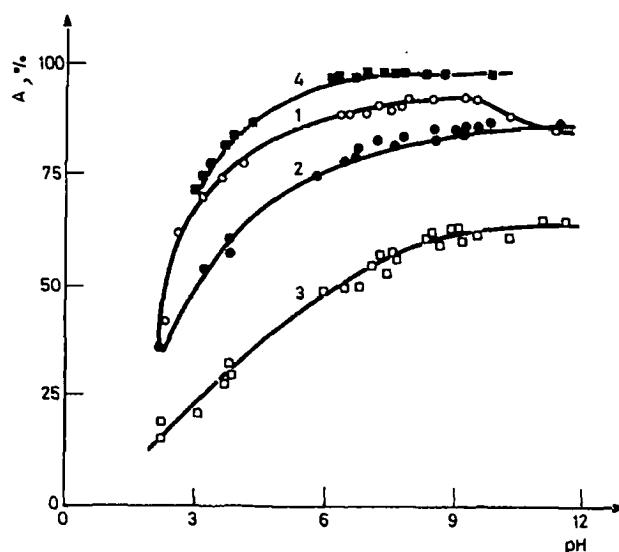


Fig. 4. Adsorption of radium (in %) on montmorillonite ($V/m = 500 \text{ cm}^3 \cdot \text{g}^{-1}$) as a function of pH and solution composition. Curves: 1 – 0.01M Na^+ , $60 \text{ mg} \cdot \text{l}^{-1} \text{ SO}_4^{2-}$; 2 – 0.01M Na^+ , $30 \text{ mg} \cdot \text{l}^{-1} \text{ Ca}^{2+}$; 3 – 0.1M Na^+ ; 4 – 0.001M Na^+

adsorption on montmorillonite significantly differs from that on kaolinite. The difference is without doubt due to different quality and/or abundance of sorption sites on both minerals. The largest part of radium is bound on montmorillonite by

negatively charged adsorption sites resulting from charge deficit formed by the substitution of Mg^{2+} , Fe^{2+} and Mn^{2+} ions for Al^{3+} in the octahedral sheets (the average charge is equal to -0.837 per unit cell) and partially also by the substitution of Al^{3+} for Si^{4+} in the tetrahedral sheets (charge -0.088 per unit cell). These sites are located both on faces and in interlayer spaces of the crystals. Only a small part of the surface charge of montmorillonite sits on the crystal edges and results from "broken bonds" and lattice defects, similarly as described for kaolinite. The ratio of both the kinds of adsorption sites (substitution and "broken bonds") on montmorillonite is apposite to that on kaolinite, which may explain the different features of the corresponding pH dependences.

The pH dependence of radium adsorption on montmorillonite in the pH region of 5–11 is determined by the effect of pH on the activity of surface sites formed from "broken bonds" (see the analogous effect for kaolinite). Because the sites play only a minor role in radium adsorption, the pH dependence is very moderate. The pronounced decrease of radium adsorption at pH less than 5 is mainly due to the competition of hydrogen ions and at pH less than 3 also due to the decomposition of montmorillonite, which is more intensive than the decomposition of kaolinite.⁸ It is interesting that the maximum decrease in radium adsorption roughly coincides with the point of zero charge of montmorillonite, which is at pH ~ 2.5 .⁹ Contrary to kaolinite, montmorillonite does not easily form aggregates with a "card-house" structure, because it has an unsuitable ratio of the length to the thickness of crystals, about 200:1. Consequently, the aggregation of montmorillonite does not seem to be a probable reason for the decrease of radium adsorption at low pH values.

Figure 4 shows the effect on radium adsorption of various ionic strength and composition of adsorption solution. The presence of sulfates again does not change the adsorption, probably because of the low concentration of sulfates used and ensuing small abundance (about 6.7%) of RaSO_4 ion pair among the dissolved forms of radium. The ionic strength exerts a rather pronounced effect on the adsorption which points to the competition between the cations added and Ra^{2+} ions in ion exchange on the surface of montmorillonite. The competition obviously depends on the charge of the competing ion. The overall effect of the electrolyte on radium adsorption is further complicated by the possible change in the surface area of montmorillonite available for the adsorption. Recent study by BOROVEC¹⁰ has shown that the concentration and composition of the electrolyte significantly affects the dispersity of montmorillonite due to its effect on the formation of sandwich-like agglomerates, tactoids.

As can be seen from Fig. 3 and Table 3, the adsorption of radium on montmorillonite strongly depends on the liquid to solid ratio, V/m. The dependence is

Table 4
Desorption (in %, after 17 hr contact) of radium from montmorillonite as a function of the pH during preliminary adsorption and of the composition of desorbing solution ($V/m=500 \text{ cm}^3 \cdot \text{g}^{-1}$ during adsorption and desorption, adsorption from the solution of 0.01M Na^+)

pH	2.6	4.2	7.1	7.6	7.8	9.2	10.0	11.2
0.1M NaCl	68.5	62.0	42.0	39.5	36.5	33.8	30.9	31.0
pH	2.2	2.9	4.0	5.4	6.8	8.1	9.2	
1.0M HCl	89.1	86.7	88.4	91.1	89.9	90.7	90.4	

similar to that found with kaolinite, but the maximum adsorption ($A_{\max}$) and the distribution coefficient K'_D are higher for montmorillonite. The latter fact may be due to the higher amount of adsorption sites on the montmorillonite. For it is known that small and thin montmorillonite crystals enable easy penetration of water and cations into interlayer spaces of montmorillonite, which represent a large "inner surface". The present experiments did not allow to distinguish whether radium is more adsorbed by the "inner" or "outer" surface of the crystals. However, by the analogy with the adsorption of UO_2^{2+} ions on the same montmorillonite¹¹ it can be assumed that both surfaces are accessible for radium. The larger adsorption of radium on montmorillonite is also in accordance with the higher cation exchange capacity of montmorillonite as compared with that of kaolinite.

The different sorption properties of montmorillonite and kaolinite towards radium are reflected also in the results of radium desorption from montmorillonite, presented in Table 4. Radium cannot be quantitatively desorbed from montmorillonite even with 1M HCl. About 10% of radium is firmly bound to some sorption sites or is adsorbed from 1M HCl. The latter possibility seems more probable, since the proportion of undesorbed radium does not depend on the pH during the preliminary adsorption. The desorption of radium with the solution of 0.1M NaCl is substantially lower, also in comparison with the analogous desorption from kaolinite. These facts reflect smaller concentration of competing cations and absence of decomposition of the adsorbent in 0.1M NaCl on one hand and a larger strength of radium bond on montmorillonite on the other hand. The dependence of the desorption on the pH during the preliminary adsorption points to possible differences in the strength of adsorption bonds formed at different pH values.

The data obtained suggest that montmorillonite may substantially affect radium adsorption on water sediments, even if present at low concentrations (several percent). Its effect will be almost independent of water pH in the pH range typical for surface

waters, but will decrease with the increasing ionic strength of water. A substantial part of radium bound by the montmorillonite component of sediments in surface waters can be released to the water phase due to an increase in the salinity of water, but the release is less easy than that from kaolinite.

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